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(54) **Grease compositions, oxyaluminum acylate intermediate compositions useful in the preparation thereof, and process for making such.**

(57) Mixed aromatic/aliphatic oxyaluminum acylates are provided which have utility for the manufacture of greases without generating by-product alcohol and without requiring the use of water and wherein the ratio of the number of aromatic radicals to the number of aliphatic radicals ranges from 2:3 to 19:1. Processes are provided for making such compounds including a continuous process. Also premix compositions useful in grease manufacture are provided along with methods for preparing new greases therefrom.

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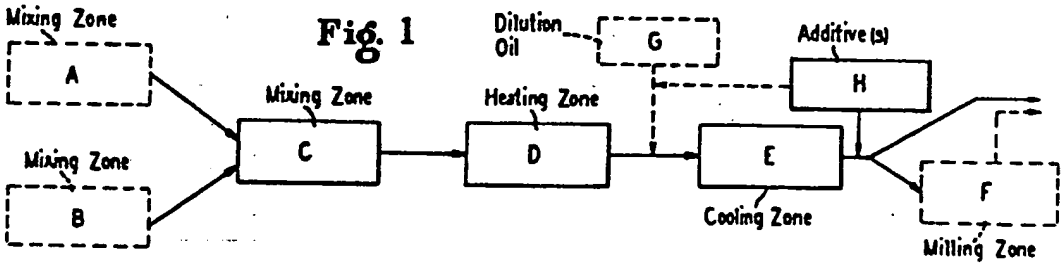
Mixing Zone
A
Mixing Zone
B
Mixing Zone
C
Heating Zone
D
Dilution Oil
G
Additive (s)
H
E
Cooling Zone
F
Milling Zone

Fig. 1

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DESCRIPTION

"GREASE COMPOSITIONS, OXYALUMINUM ACYLATE INTERMEDIATE COMPOSITIONS USEFUL IN THE PREPARATION THEREOF, AND PROCESS FOR MAKING SUCH"

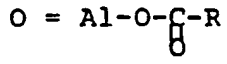
BACKGROUND

Aluminum complex greases incorporate an aluminum complex soap which is an hydroxy aluminum diacylate wherein the acylates differ from one another. Conventionally these soaps are made in situ; see U.S. Patent 3,345,291, U.S. Patent 3,054,816 and U.S. Patent 3,776,846. All these prior art techniques involve alcohol and possibly also water removal.

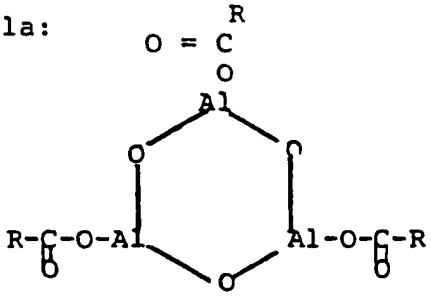
Such removal problems are avoided by using a new class of mixed oxyaluminum acylates which can be prepared by a three acid route, a two acid route, or a controlled hydrolysis route, as explained hereinbelow. This new class differs from prior art efforts to make greases with oxyaluminum acylate because this new class contains a higher amount of aromatic acid which unexpectedly helps the grease making procedure.

BRIEF SUMMARY OF THE INVENTION

In one aspect, the present invention is directed to a class of new and very useful mixed aromatic/aliphatic oxyaluminum acylates representable either by the formula:

(1) 

or by the formula:

(2) 

wherein R is selected from the group of radicals consisting of Type (A) radicals and Type (B) radicals where:

Type (A) radicals consist essentially of aliphatic radicals each containing from 15 to 38 carbon atoms, and

Type (B) radicals consist essentially of aromatic radicals each containing from 6 to 16 carbon atoms.

Also, in any given group of such formula (1) and/or formula (2) compounds, the ratio of the number of radicals of said Type (B) radicals to said Type (A) radicals ranges from 2:3 to 19:1.

In another aspect, the present invention relates to a process for making compounds of formulas (1) and (2) which contain up to 75 mole percent of aromatic carboxylic acid by utilizing three carboxylic acids: An aromatic acid, an aliphatic acid, and a lower alkanoic acid, the latter producing during oxyaluminum acylate manufacture an ester by-product which is easily volatilized and removed.

In another aspect, the present invention relates to a process for making compounds of Formulas (1) and (2) by utilizing two carboxylic acids: An aromatic acid and an aliphatic acid.

Thus, the present invention in one aspect provides a process for making a composition comprised on a 100 mole percent basis of about 50 mole percent of mixed oxyaluminum acylate with the balance up to 100 mole percent thereof being mixed esters of carboxylic acid material. Not only does such a product composition appear to avoid certain herein indicated problems associated with mixed oxyaluminum acylates made by the three acid route, but also and surprisingly the product compositions appear to be readily dispersible (including solubilizable) in organic liquids of the type conventionally used in making greases. It is theorized

that the presence of mixed esters of carboxylic acid material in combination with the mixed oxyaluminum acylate product for some reason not now clear promotes the dispersibility and solubilization of the mixed oxyaluminum acylate in such organic liquid (there being no intent herein to be bound by theory).

In another aspect, the present invention relates to premix compositions for use in grease manufacture which compositions comprise on a 100 weight percent total weight basis:

(a) from about 30 to 70 weight percent of at least one group of compounds of this invention as defined above, and, correspondingly

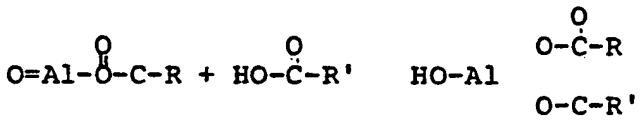
(b) from about 70 to 30 weight percent of a petroleum derived hydrocarbon liquid having a viscosity at 100°F ranging from about 35 to 50,000 SUS or of any other suitable liquid which would be compatible with grease systems such as synthetic oil or ester of the type conventionally used and/or known to be compatble with synthetic lubricating oil systems.

In such a composition, the above indicated component (a) is uniformly dispersed in the above indicated component (b). As used herein, the term "dispersed", "dispersion", or the like is inclusive of both solutions and suspensions. Preferably, such a composition of this invention has the component (a) substantially completely dissolved in the component (b). This aspect further provides methods for the preparation of such compositions. It is noted that the terms "component (A)" and "component (B)" used herein are different from the terms "Type (A) radicals" and "Type (B) radicals" and should not be confused with each other.

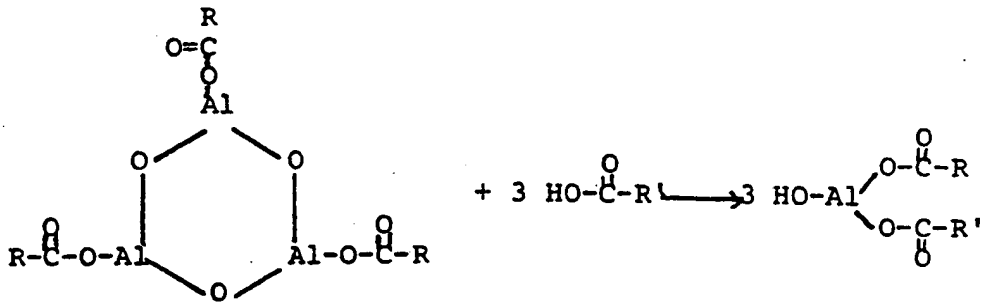
In another aspect, this invention relates to an improved process for making a grease. This process involves

the step of converting compounds of this invention as above defined in Formulas (1) and/or (2) which are dispersed (preferably dissolved) in an oil (preferably a petroleum derived hydrocarbon oil) by reaction with a carboxylic acid material into a hydroxyaluminum diacyl soap directly without the production of by-product alcohol and without water being present. The following chemical equations are illustrative of this addition reaction whereby no by-products are formed:

Equation I where the compounds of this invention are represented by Formula (1)



Equation II where the compounds of this invention are represented by Formula (2)



In such equations I and II, and below, R is as above defined, and R' is a radical supplied from among the usual carboxylic acids added by grease manufacturers practicing this process. For example, R' can preferably be the same as R except that the ratio, in any given instance, of Type (B) radicals to said Type (A) radicals can range from 0 to about 5:1. For example, by one presently preferred procedure of this invention, this process involves the steps of heating a mixture of a

group of compounds of this invention as above defined in Formulas (1) and (2) with a petroleum derived hydrocarbon having a viscosity at 100°F ranging from about 35 to 50,000 SUS (though higher and lower viscosity oils may be used if desired) until substantially completely dissolved in such petroleum oil. Next, to such resulting solution is added at least one carboxylic material selected from the group consisting of aliphatic carboxylic acids, aromatic carboxylic acids and mixtures thereof, as more particularly hereinbelow defined. Thereafter, the temperature of the resulting system is raised and maintained at some elevated temperature until at least some of the compounds of this invention present in the system are converted to hydroxyaluminum diacylate soap through reaction with the acids added thereto. In another modification of this invention, the carboxylic acids can be added to the lubricating oil base first and then the compounds of this invention added to the resulting system.

In another aspect, this invention is directed to methods for making improved grease by a process step sequence wherein an aluminum alkoxide is first converted to a mixed oxyaluminum acylate in combination with mixed esters of carboxylic acid material after which such system is subject to reaction with carboxylic acids in a grease making liquid to produce a product grease.

In another aspect, this invention relates to improved greases produced by such grease making process of the present invention. In making such an improved grease, the carboxylic acids can be added to the starting oil base first and then compound(s) of this invention added to this resulting system, or otherwise, if desired.

In another aspect, this invention relates to improved continuous process for making aluminum complex greases.

A principal feature of the present invention is the

creation of smooth, clear greases in a simple and reliable manner. No special pains are needed to predissolve the benzoic acid (e.g. aromatic acid) used with the compounds of this invention as above defined in formulas (1) and (2) and no special sequential addition and treating is needed. Both the aromatic acid (e.g. benzoic acid) and the aliphatic acid (e.g. fatty external acids) can be added simultaneously as a solid powder mixture, if desired, a smooth, clear grease is characteristically obtained.

Another principal feature of the present invention is that such mixed aromatic oxyaluminum acylates (as defined hereinabove) permit one to prepare a grease having excellent and controllable high viscosity characteristics compared to the prior art (see, for example, Rinse U.S.P. No. 3,054,816).

Another feature of the present invention is that such aluminum acylates (as defined hereinabove) permit one to prepare a grease without the use of added water and without the production of any by-product alcohol whatsoever. The freedom from by-product alcohol formation is highly desirable both from an environmental standpoint and also from a process operation standpoint.

The grease products of this invention characteristically incorporate an aluminum complex soap which, as those skilled in the art appreciate, has reference to a mixture of aluminum soap molecules containing at least one hydroxyl anion for each aluminum cation and substantially two carboxylic acid anions per aluminum atom. By this invention, such an aluminum complex soap has two dissimilar acid anions, such as one aromatic (e.g. benzoate anion) and one saturated aliphatic (e.g. arachidate, stearate, or like fatty carboxylic acid anion). Such an aluminum complex soap is produced by chemical reaction with the mixed oxyaluminum acylates of this invention when the same are used to make a grease in accordance with

teachings of the present invention. Specifically, such an aluminum complex soap is generated in situ in a mineral oil continuous phase during the practice of the grease making process of this invention through reaction with carboxylic acid material, but unlike the in situ process of U.S.P. No. 3,345,291, no by-product alcohol is produced.

Characteristically, a controllable and uniform thickening of a starting petroleum composition is achieved by the practice of the process of the present invention using the mixed oxyaluminum acylates of this invention.

Other further objects, aims, purposes, features, advantages, uses and the like will be apparent to those skilled in the art from the present disclosure.

BRIEF DESCRIPTION OF DRAWINGS

Figure 1 is a simplified flow sheet illustrating one embodiment of the present invention; and

Figure 2 is a flow sheet illustrating a further embodiment of the present invention.

DETAILED DESCRIPTION

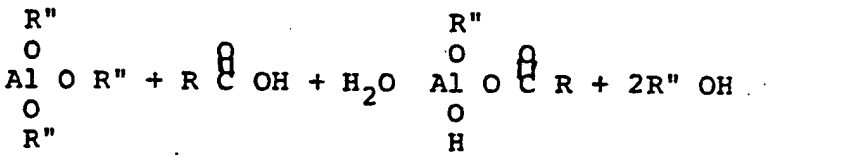
As those skilled in the art will appreciate, oxyaluminum acylates of which the mixed oxyaluminum acylates of this invention, as defined above in Formulas (1) and (2) are examples, are believed presently to exist either in a monomeric form or in a cyclic trimeric form. The conditions under which one form exists as opposed to the other form are at this time completely unknown.

One class of preferred compounds of this invention are those wherein the Type (B) radicals are derived from benzoic acid. Another class of preferred compounds of this invention are those wherein the Type (A) radicals are derived from stearic acid or isostearic acid and wherein the Type (B) radicals are derived from benzoic acid.

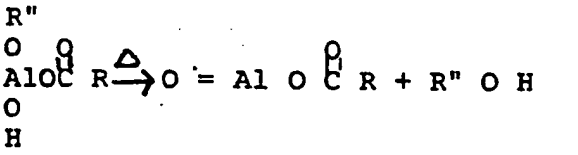
One class of preferred aliphatic carboxylic acids for this invention comprises acids derived from fish oil which contains at least about 50 percent by weight of hydrogenated fatty acids of arachidic and behenic acids, such as "Hydrofol 2022-55", available from the Ashland Chemical Company of Columbus, Ohio, USA.

Any convenient method of making the mixed oxyaluminum acylates of Formulas (1) and/or (2) may be employed including the controlled hydrolysis method where approximately one mole of total acids and approximately one mole of water are reacted with approximately one mole of an aluminum lower alkoxide. The following equations represent the reaction:

Equation III



Equation IV

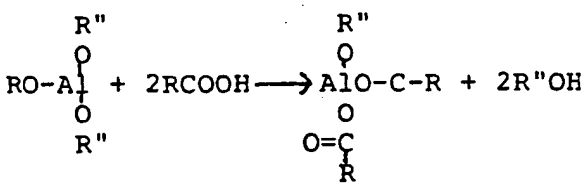


where R is as defined above and R" is a lower alkyl group. Equations III and IV show the reactions of preparing compounds represented by Formula (1). As those skilled in the art will appreciate, similar equations showing the preparation of compounds represented by Formula (2) can be developed by starting with three moles aluminum lower alkoxide, three moles of acid and three moles of water.

The desired number ratio of aliphatic radicals to aromatic radicals is achieved by controlling the composition of RCOOH in Equation III and/or IV.

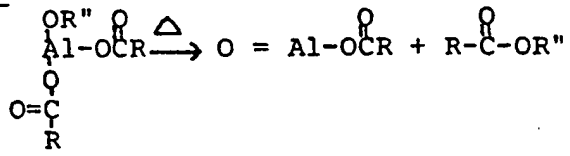
A basic method of preparation involves a two step procedure. Thus, in a first step, two moles of carboxylic acid are combined with one mole of an aluminum trialkoxide (e.g. from about 82 to 150°C) and reacted at an elevated temperature to form an aluminum alkoxy diacylate and two moles of an alcohol by-product (which is continuously distilled off) as illustrated by the following equation:

Equation V



wherein R is defined above in relation to Formulas (1) and (2) and R" is as defined above. Then, in second step, the product aluminum alkoxy diacylate is thermally decomposed conveniently at atmospheric pressures using, for example, a temperature of from about 150 to 250°C. The thermal decomposition results in a splitting off of an ester composed of the remaining (-OR") group and one of the acylate groups, as illustrated by the following equation wherein compounds of formula (1) are formed:

Equation VI



Equation VI is illustrative of the reaction of preparing compounds of formula (1).

When making a long chain oxyaluminum acylate, where R is derived from stearic acid, for example, the by-product

ester is a high boiling acid which is removed by vacuum distillation. In a preferred procedure, however, the intermediate aluminum monoalkoxide diacylate is a mixed diacylate, where one of the acyl groups is a low molecular weight alkanoic acid. The ester is then formed with the acyl group of the lower molecular weight acid as the by-product, and this ester can be distilled off easily at atmospheric pressures. Thus, the higher molecular weight fatty acylate group is left behind to form the oxyaluminum acyles. Although Rinse in U.S. Patent 2,948,743 of August 9, 1960 shows using acetic acid with stearic acid, this patent does not teach or suggest the preparation of the mixed oxy aluminum acylates of this invention. Acetic acid is presently preferred lower alkanoic acid herein because of the characteristically low boiling points associated with acetate esters (isopropyl acetate, for example, boils at 88.4°C).

The term "lower" as used herein has reference to a molecule or radical, as the reference may be, containing less than five carbon atoms per molecule.

This process of the present invention thus comprises the use of at least three acids, one of which is aromatic, and all three acids are add mixed together and reacted simultaneously with the aluminum tri-alkoxide. The amount of lower alkanoic (i.e. acetic) acid in such mixture is equal to one mole per mole of aluminum, and the other acids in the mixture together are equivalent to one mole mixed acids per mole of aluminum.

In compounds of this invention made by methods where a lower alkanoic acid such as acetic acid is used to produce a volatile ester by-product, it is theorized (although not herein bound by theory) that minute amounts (1% or less) of the oxyaluminum lower alkanoate (such as oxyaluminum acetate)

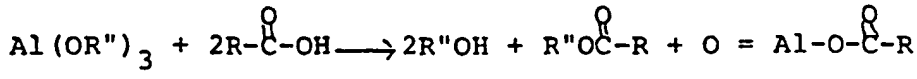
could be present. However, based on present available knowledge, these small quantities of such oxyaluminum lower alkanoate do not appear to adversely affect the properties of greases made from the aforesaid compounds of this invention.

A reaction sequence used to make a compound of this invention (see Equation III and/or Equation IV) can be carried out in organic liquid phase or in some cases it can be carried out as a mass reaction ("neat"). Particularly when it is desired to use the product mixed oxyaluminum acylate in grease making (as herein described and illustrated), it is preferred, but not necessary, to conduct the synthesis in a mineral oil or ester of the type which approximates that which it is anticipated will be used subsequently for an actual grease making operation. Then, the product as synthesized can be used directly for grease making without further preparative procedures.

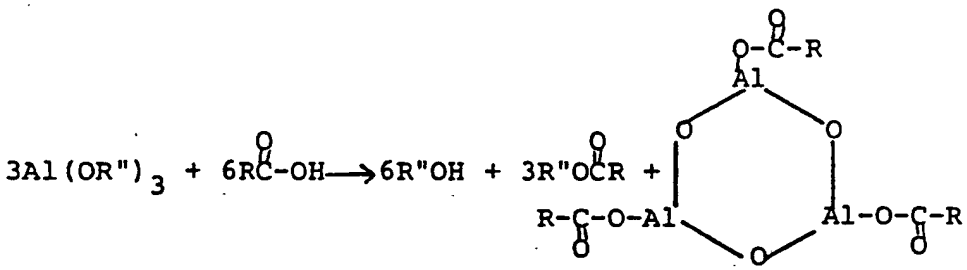
However, when, for example, it is desired to make such a mixed oxyaluminum acylate of Formula (1) and/or (2) in a purified or concentrated form, then the synthesis reaction(s) can be conducted in some cases without solvent or in a relatively low boiling organic inert (as respects reaction products) solvent. Afterwards (if conducted in a solvent) this solvent can be removed by vacuum distillation preferably at reduced pressures to leave a purified, concentrated product. When, for example, such a concentrated product comprised of a compound of this invention is to be used for viscosity regulation of a liquid curable polyester resin system in a vinyl monomer, such as styrene, such concentrated product can be dissolved in such styrene and the resulting solution then added to the polyester resin in a desired amount.

A preferred preparation procedure involves the reaction represented by the following equations:

Equation VII



Equation VIII



In the above equations: R is as above defined; R" is a lower alkyl radical. Equation VII shows the reaction of preparing compounds represented by Formula (1) and Equation VIII shows the reaction of preparing compounds represented by Formula (2). The desired number ratio of aliphatic radicals to aromatic radicals is achieved by controlling the composition of RCOOH in Equation VII and/or VIII.

This preferred method of preparation involves a two step procedure. Thus, in a first step, two moles of carboxylic acid are combined with one mole of an aluminum trialkoxide (e.g. from about 65 to 150°C) and reacted at an elevated temperature to form an aluminum alkoxy diacylate and two moles of an alochol by-product (which continuously distilled off) as illustrated by Equation V above.

Then, in second step, the product aluminum alkoxy diacylate is thermally decomposed conveniently at atmospheric pressures using, for example, a temperature of from about 150 to 250°C. The thermal decomposition results in a splitting off

of a by-product ester composed of the remaining (-OR") group and one of the acylate groups, as illustrated by the equation VI above, wherein compounds of Formula (1) are formed:

When making a mixed oxyaluminum acylate of the present invention, a convenient method to use as a starting carboxylic acid mixture, is a mixture of the two acids which will supply the desired radicals for the said mixed oxyaluminum acylate. When, for example, an oxyaluminum benzoate/stearate of the type disclosed by the present invention is desired, the acid mixture would be conveniently comprised of benzoic acid and stearic acid. In the above case, the ester formed by the reaction would be a mixture comprised of benzoate and stearate esters. These esters or a portion of them can be removed by vacuum distillation or can be left with the oxyaluminum acylate as a carrier fluid.

In a premix composition, of the invention, presently preferred inert organic liquids include a petroleum derived hydrocarbon, a phthalate ester, esters produced as by-products in the making of oxyaluminum acylates of this invention, and mixtures thereof. By the "inert organic" in reference to "liquid" reference is had to the circumstance that such liquid is itself not reactive with oxyaluminum acylates under the conditions of manufacture and use described herein.

Any convenient preparation method, as described herein, may be employed to make a premix composition of this invention useful in grease making. One presently preferred method, for example, involves, as a first step, admixing of at least one aluminum trialkoxide with a mineral oil which has a viscosity at 100°F of from about 35 to 50,000 SUS and which has dispersed therein from about 30 to 72 weight percent of a carboxylic acid mixture based upon the total combined weight of such compounds and said mineral oil.

To make a grease of this invention using a mixed oxyaluminum acylate of Formula (1) and/or (2), for example, one employs a mineral starting oil having a viscosity at 100°F of from about 35 to 50,000 SUS. In such oil, at least one carboxylic acid material is contacted with such mixed oxyalumimnum acylate with preferably both reactant types being dispersed (more preferably dissolved) in the oil. Such contacting carried out at a temperature sufficient to produce reaction between said carboxylic acid material and said oxyaluminum acylate compound, and such contacting is continued until at least some of such oxyaluminum acylate compound has been converted into an aluminum soap. The product aluminum soap is an hydroxy aluminum diacylate. The resulting grease containing such hydroxy aluminum diacylate is then milled and packaged. It can be milled at room temperatures or at any elevated temperatures up to about 200°C with temperatures below about 150°C being presently preferred.

In one presently preferred grease making grease process of the present invention, the following steps are employed:

First, one heats mixture of petroleum derived hydrocarbon oil having a viscosity of 100°F of from about 35 to 50,000 SUS and a grease making composition as above described. This mixture contains a total amount of aluminum in the range from about 0.01 to 2.0 weight percent based on total mixture weight. Such heating is conducted at temperatures, and for times, sufficient to substantially completely dissolve or uniformly disperse all starting mixed oxyaluminum acylates present in said hydrocarbon oil.

Next, one admixes with the resultant such mixture of Step One a total of from about 0.8 to 1.2 moles (per mole of oxyaluminum acylate present in said resultant such mixture) of at least one carboxylic acid material selected from the group

consisting of aliphatic monocarboxylic acids containing from 15 through 40 carbon atoms each and aromatic monocarboxylic acids containing from 6 through 16 carbon atoms each.

Finally, one heats and gradually raises the temperatures of the product mixture, all the while agitating such product mixture, until at least some of such starting mixed oxyaluminum acylates present in the first step have been converted into hydroxy aluminum diacylate (aluminum soap) by reaction in situ with said carboxylic acid material.

As indicated, in such grease making process of this invention, the starting mixed oxyaluminum acylates of Formula (1) and/or Formula (2) present in a base oil are reacted preferably substantially completely with carboxylic acid materials. A starting such mixed oxyaluminum aculate provides from a stoichiometric standpoint approximately one-half of the acylate radicals needed to produce an aluminum soap which is formed from the reaction of such mixed oxyaluminum acylate with carboxylic acid material, such aluminum soap being a compound which contain approximately two acyl groups and one hydroxyl group each group being directly bonded to an aluminum atom (one name for such soap being hydroxy aluminum diacylate).

In calculating the molar quantity of carboxylic acid material to be used (added) for reaction with a mixed oxyaluminum acylate in making a grease according to this invention (based on the number of carboxyl groups present in the carboxyl acid material), it is sometimes convenient to use a mole ratio ranging from about 0.8 to 1.2 of total quantity of carboxylic acid material to total quantity of mixed oxyaluminum acylate.

In a grease prepared by the teachings of this invention, such an aluminum soap is preferably characterized by having the total number of acyl radicals of any given soap molecule composed of a ratio of aliphatic acyl groups to

aromatic acyl groups ranging from about 1.3:0.7 to 0.7:1.3. Presently preferred aliphatic acyl groups are derived from fatty carboxylic acids each mixture containing an aliphatic group of at least about 16 carbon atoms. Also, presently preferred aromatic acyl groups are derived from benzoic acid.

In a grease prepared by the teachings of this invention, it is not necessary to have all of the starting mixed oxy-aluminum acylate compounds converted to such an aluminum soap.

In addition to, or in admixture with, petroleum derived (mineral) grease making base oils, suitable specialized starting oils adapted for use in the grease making process of the present invention include lubricating oils of naphthenic base, paraffinic base hydrocarbons, mixed base mineral oils, vegetable oils, synthetic oils, including synthesized hydrocarbon base fluids, alkylene polymers, polysiloxanes, ester-type oils such as dicarboxylic acid ester type oils, liquid esters of phosphorous acids, (such as are shown in U.S.P. No. 2,768,138), and the like. In general, preferred starting base oils have viscosities at 100°F ranging from about 35 to 50,000 SUS, but other inert organic liquids can be used with viscosities outside of the range.

To make a grease using compounds of Formula (1) and/or (2) in an oil, a grease maker need use no particular type of carboxylic acid material for reaction therewith. For example, it now appears that the teachings of the prior art with respect to the use of various carboxylic acids, combinations thereof, order of contacting, temperature conditions, and the like in connection with the use of the prior art aluminum alkoxides in grease making can be employed to make greases from compounds of Formula (1) and/or (2), except that here no by-product alcohol is produced and no water is needed. Mono and dicarboxylic acids can be used, as can halo substi-

tuted such acids like chloroacetic acid, dichloroacetic acid, and the like. Examples of suitable dicarboxylic acids include succinic. One particularly preferred monocarboxylic acid is presently isostearic because such acid, which is a branched C_{18} saturated acid, is a relatively low viscosity liquid at ambient conditions and tends to bring down the melting point and softening point of derivatives thereof, including especially aluminum soaps thereof. Dimer acids, such as dimerized vegetable oil carboxylic acids, such as are offered commercially by Emery Industries, can also be used as the carboxylic acid material.

In such class, the Type (B) radicals are preferably derived from benzoic acid. Such preferred compound is relatively easy for a grease maker to convert into a grease in the presence for example, of a hydrocarbon oil. Such compounds containing a higher ratio of benzoic acids to aliphatic acids than is disclosed in the prior art presently appear to be particularly desirable in grease making because a smaller quantity of benzoic acid is subsequently needed to complete the in situ reaction which forms the hydroxy aluminum stearate/benzoate soap. Benzoic acid itself is difficult for a grease maker to handle because of its tendency to sublime at temperatures above 100°C.

Greases made with mixed oxyaluminum acylates as provided by the teachings of this invention can be formulated with the various additives heretofore employed in the grease making art, if desired.

To prepare a composition containing about 50 mole percent mixed oxyaluminum acylate, with the balance up to 100 mole percent being mixed esters of carboxylic acid material, using the two acid preparation route, one heats a mixture of an aluminum alkoxide material and a carboxylic acid material to a first temperature which is above the melting

point of the carboxylic acid material, but which is below the boiling point of an alkanol material while agitating the mixture. Such first heating is continued for a time sufficient to form a substantially single phase liquid or a substantially uniform slurry comprised of such mixture.

In such mixture the aluminum alkoxide material is characterized by the formula:

(3) $Al(OR")_3$

where R" is as defined above.

Also, in such mixture, the carboxylic acid material is comprised of:

(a) at least one aliphatic monocarboxylic acid containing from 8 to 40 carbon atoms per molecule,

(b) at least one aromatic monocarboxylic acid containing from 7 to 17 carbon atoms per molecule, and

(c) the mole ratio of said aromatic acid to said aliphatic acid ranging from about 2:3 to 19:1.

Also, such alkanol material is characterized by the formula:

(4) $R"OH$

where R" is as defined above and where R"OH is derived from reaction between said aluminum alkoxide material and said carboxylic acid material.

In such first heating, such mixture is characterized by having a mole ratio of said aluminum alkoxide material to said carboxylic acid material of about 1 to 2.

Preferably, before said first heating, said carboxylic acid material is heated to such first temperature and the

aluminum alkoxide material is then admixed therewith.

After the desired single phase liquid or a substantially uniform slurry is produced by such first heating, a second heating is undertaken, preferably without any cooling. The second heating is preferably conducted with continuous agitation of the liquid, and during the second heating the liquid is heated to second temperatures in the range where such alkanol material is distilled off. The second heating is maintained or continued for a time sufficient to remove the equivalent of at least about two theoretical moles of said alkanol (per mole of such aluminum alkoxide material).

Next, and preferably without cooling, the product from such second heating, a third heating is undertaken. In the third heating, the resulting product from the second heating is gradually heated to third temperatures ranging from a temperature corresponding to the final temperature of the second heating up to a temperature of about 200°C, thereby converting the resulting product from the second heating into a homogenous liquid which has characteristically a viscosity which is substantially less than that of the product resulting from the second heating.

Preferably, the third heating is continued at such 200°C for a time of at least about 1/4 hour, and more preferably such third heating is continued at such 200°C for a time of at least about 0.5 hour.

In one presently preferred procedure, after such third heating, the final liquid so produced is subjected to a vacuum distillation so as to remove from such final liquid any organic material therein which at atmospheric temperature and pressure boils at a temperature not more than about 250°C. Preferably, such a vacuum distillation is conducted at a pressure not above about 700 mm Hg. At such a reduced pressure

it is preferred to keep the distillation temperatures below about 200°C.

During the second heating, it is theorized that the reaction which occurs is represented by Equation V above.

In the third heating step, it is presently theorized that a reaction occurs which is representable by Equation VI above.

By the present invention, there is provided a simple, reliable, economical continuous process for making an aluminum complex grease wherein a sequence of steps is practiced simulataneously but sequentially. In a first step, one mixes a first dispersion of oxyaluminum acylate material with a second dispersion of carboxylic acid material in a first mixing zone to produce a substantially homogenous product mixture. Preferably, such dispersions are true solutions.

In the next steps, one charges such product mixture into an elongated reaction zone, and then passes such product mixture through such reaction zone while maintaining said reaction zone at a temperature ranging from about 135 to 250°C. The residency time for such product mixture in such reaction zone is at least such that the oxyaluminum acylate material is substantially completely reacted with the carboxylic acid material, thereby to form a product aluminum complex grease.

Thereafter, one removes such product grease from such reaction zone, and cools same.

After a preliminary cooling, a product grease can be milled, if desired, or mixed with additional oil and additives, if desired, in one or more subsequent mixing zones.

The oxyaluminum acylate material for use in this continuous process is as defined above in formulas (1) and (2), with references as above shown.

Similarly, the carboxylic acid material utilized in this continuous process is characterized by the formula:

(5) $R'-\overset{O}{\overset{\|}{C}}-OH$

where R' is selected from the group consisting of aliphatic radicals containing from 9 through 40 carbon atoms each and aromatic radicals containing from 6 through 28 carbon atoms each, and wherein, in any given such carboxylic acid material, the weight percent of said aromatic radicals ranges from and including 0 to about 70% with the balance up to 100 weight percent of all said radicals, being said aliphatic radicals.

The carrier liquid for each of said first and said second dispersions used in this continuous process comprises an inert organic liquid, preferably one having a viscosity at 100°F of from about 35 to 50,000 SUS, more preferably from about 50 to 25,000 SUS, such as a hydrocarbon oil.

The total amount of aluminum in such product mixture used in this continuous process ranges from about 0.01 to 4.0 weight percent based on total mixture weight, and preferably from about 0.01 to 2 weight percent.

The mole ratio of the total amount of aluminum to the added said carboxylic acid material used in this continuous process ranges from about 0.75:1.25 to 1.25:0.75 in said product mixture, and preferably ranges from about 0.8:1.2 to 1.2:0.8.

One preferred class of first dispersions for use in the practice of such continuous process comprises on a 100 weight percent total weight basis:

(A) from about 2 to 60 weight percent of oxy-aluminum acylate material as defined above, and, correspondingly

(B) from about 98 to 40 weight percent of a petroleum derived hydrocarbon liquid having a viscosity at 100°F ranging from about 35 to 50,000 SUS.

In the invention, in place of, or in combination with, such hydrocarbon liquid, one can employ any other hydrocarbonaceous liquid which would be compatible with grease systems, such as a synthetic oil, an ester of the type conventionally used or known to be compatible with synthetic lubricating oil systems, or the like. In such a first dispersion, the above indicated component (A) is uniformly dispersed in the above indicated component (B). Although, as indicated above, the term "dispersed", "dispersion", or the like as used herein, is inclusive of both solutions and suspensions, preferably, such a dispersion used in the continuous process has the components (A) substantially completely dissolved in the component (B). Conventional methods for the preparation of such dispersions can be employed.

One preferred class of second dispersions for use in the practice of the continuous process comprises on a 100 weight percent total weight basis:

(A) from about 2 to 50 weight percent of carboxylic acid material and, correspondingly

(B) from about 50 to 98 weight percent of a petroleum derived hydrocarbon liquid having a viscosity at 100°F ranging from about 35 to 50,000 SUS.

The continuous process of this invention involves the converting of compounds as defined in Formulas (1) and/or (2) which are dispersed (preferably dissolved) in an oil (preferably a petroleum derived hydrocarbon oil as described above) by reaction with a carboxylic acid material as defined in Formula (3) into an hydroxyaluminum diacyl soap directly with-

out the production of by-product alcohol and without water being present as illustrated by the equations above presented (see, for examples, equations I and II).

Any convenient method may be used to make starting solutions or dispersions for use in the continuous process of the present invention which features the continuous production of smooth, clear greases in a simple and reliable manner.

The continuous process of this invention is advantageously and preferably practiced with a first dispersion which additionally contains an ester material having the formula:

(6) $RC(=O)-OR''$

wherein

R is as defined above, and

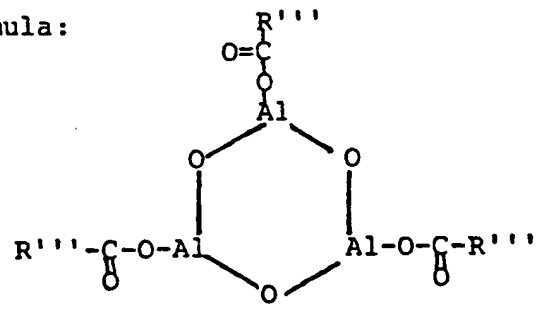
R" is a lower alkyl radical, and wherein, in said first dispersion, the mole ratio of said ester material to said oxyaluminum acylate material ranges from about 1.5:1 to 0.

Further, the continuous process of this invention is preferably and advantageously practiced by utilizing a first dispersion which additionally contains, or even comprises, an oxyaluminum acylate material which is characterized by the formula:

(7) $O = Al-O-C(=O)-R'''$

and by the formula:

(8)



wherein R''' is as R is defined above but additionally includes a further group of radicals consisting of:

Type (C): lower alkyl radicals, and

wherein, in any given such oxyaluminum acylate material on a 100 mole percent total basis, the mole percent of said Type (B) radical ranges from about 40 to 95, the mole percent of said Type (C) radicals ranges from 0 to about 50, and the mole percent of said Type (A) radicals ranges from about 5 to 50.

Preferably, a first dispersion for use in the continuous process is prepared inherently in a process for making oxyaluminum acylates as described above.

Conveniently and preferably, in practicing the continuous process each of the first dispersion and the second dispersion can be heated to temperatures which facilitate pumping, dissolution of the respective ingredients, and mixing. The respective first and second dispersion can each be pumped by a metered pump into a mixer of some desired but conventional type, such as a static mixer, a Welex-type high intensity mixer, Prodex mixer, or the like. The mixed product stream coming from such a mixing zone then passes either directly to a reaction zone or to a heat exchange zone. If a separate heat exchange zone is employed, the temperature of the product mixture is raised to a temperature compatible with the temperatures maintainable in the reaction zone. In the reaction zone, the product mixture is heated to a temperature in the range from about 135 to 250°C (preferably about 200°C) in order to cause the oxyaluminum acylate material to react with the carboxylic acid material and thereby form a grease.

Observe that the continuous process of this invention from a chemical engineering point of view is relatively simple, requiring at a minimum two feed tanks, a mixing system of some kind (perhaps a static mixer for simplicity), a

long pipe or equivalent about which a heat exchange medium can be passed so as to serve as a reaction zone. The length of the tubular reaction zone is determined by the rate at which the material is pumped through the reaction zone and how long the time period is for heating the product mixture to a reaction temperature. Production capacity appears to depend upon the rate at which mixtures can be pumped through the system using available heat transfer capacity.

After passing through the continuous reaction zone, the product grease is cooled, optionally milled in whole or in part, and optionally blended in whole or in part with various grease additives.

A present preference in the continuous process is to entirely cool a process grease to a lower temperature before subjecting all or a part of a product grease to a milling operation; however, if one desires to mill an entire grease, the entire product grease can be pumped through the milling zone. Conventional milling equipment may be employed. However, if one only wishes to mill a fraction of a grease product, a side stream of product grease can be diverted for the main stream, milled, and recombined with the continuously flowing main stream.

One feature of the continuous process is that milling is not required for certain greases, it appears, since there is little opportunity for the continuous processing system to develop localized hot spots which, it is theorized, may well be the main cause of grease lumping. It is the phenomenon of grease lumping which has heretofore led to the desirability of milling, as those skilled in the grease making art will readily appreciate.

In addition to, or separately from, a milling zone, if desired in the continuous process one can also meter in and mix with a product grease various additives in appropriate

mixing zones. The additives can be added to a product grease either before or after a milling operation, as those skilled in the art will appreciate. Mixing of a product grease with additives may be accomplished conventionally.

In the case of a tubular reactor, in order to avoid irregular heating, such as might occur in the region adjacent the reactor wall, a mixing of reactants in the reactor is sometimes desirable. For this purpose, a modified extruder or a static mixer may be employed in the reaction zone.

In one presently preferred method of operating the continuous process, after a product grease is removed from the reaction zone, the product grease may be cooled in a heat exchanger to a temperature ranging from about 140 to 25°C before being milled, when milling is utilized. To avoid the use of a heat exchanger, it may be convenient to withhold a large portion of oil needed for the finished grease and add it to the system after the aluminum containing product grease passes through the reaction zone.

In one presently preferred mode of practicing the continuous process, after cooling and before milling, additives may be mixed with a product grease in a continuous mixing zone interposed in the system before the milling zone.

If desired, only a portion of a product grease, so cooled as above-described and so milled as above-described, may be mixed with the remainder of the product grease under continuous operating conditions, wherein cooling and milling are accomplished using only a portion of a given product grease. After the milling, the portion is conveniently mixed with the remainder of a product grease in a third mixing zone. If desired, various additives may be mixed with the product grease in such third mixing zone.

For reasons of process simplicity, a present preference is to continuously process an entire product grease by a cooling and milling operation so as to avoid side streams. If desired, after cooling and before milling, additives may be mixed with a product grease in such a second mixing zone and further additives may be mixed with such a product grease in such a third mixing zone.

Conveniently, and preferably, in practicing the continuous process of the present invention, the total amount of carrier liquid present in the first mixing zone is derived from the first dispersion and the second dispersion. A present preference is to maintain the weight ratio of carrier liquid in said first dispersion to the weight of carrier liquid in the second dispersion within a weight ratio ranging from about 80:20 to 20:80 (based upon the total weight of carrier liquid present in the first mixing zone).

Preferably in the first mixing zone, the total amount of aluminum present ranges from 0.01 to 4.0 weight percent of the total product mixture on a 100 weight percent basis.

If desired, a plurality of first make-up tanks may be employed, and more than one first feed tank may be employed.

Referring to Figure 1, there is seen a simplified flow diagram of one mode of practicing the continuous process of the present invention. In first mixing zone A, one can prepare a solution of oxyaluminum acylate of the type hereinabove defined, while in a second mixing zone B one can prepare a solution of monocarboxylic acids as above defined. The solutions from the respective mixing zones A and B are then charged into another mixing zone C, in proportions so as to produce a product mixture as

hereinabove defined. From the mixing zone C, the product mixture is moved into a heating zone D wherein the oxy-aluminum acylate material reacts with the carboxylic acid material to produce an aluminum soap as hereinabove described with the result that the material issuing from the heating zone D constitutes a grease product of the present invention.

The grease product from heating zone D, if desired, can optionally be admixed with dilution oil from a holding tank G or the like, the dilution oil and the product grease after heating zone D, it is preferred to charge additives into the dilution oil as such is being admixed with the product grease. Addition of additives is a common practice in the manufacture of aluminum complex greases, as those skilled in the art will appreciate, and the additives typically employed in the manufacture of aluminum complex greases are here contemplated in the amounts conventionally employed.

The product grease from heating zone D moves into a cooling zone E. The cooling zone E can be in the nature of a heat exchanger, for example. Alternatively, the cooling zone E can comprise a mixing zone wherein dilution oil at ambient temperatures is admixed with the heated product grease from the heating zone D.

Following the cooling zone E, additives may be added to a product grease, the amount of cooling occurring in a cooling zone being such as to reduce the temperature of a product grease to a level above ambient where additives are conveniently mixed with a product grease, particularly when the additives involved should not be added for reasons of product stability or the like to the grease issuing from heating zone D. Typically, additives are added to a product grease issuing from cooling zone E and such additives are admixed with a product grease in a mixing zone,

not detailed in Figure 1. The product from the cooling zone, typically with additives as indicated, can then be moved into a storage area, containerized, or the like, as desired, as as those skilled in the art will appreciate. Alternatively, the product from cooling zone E can be moved into a milling zone so as to mill a product grease to some predetermined desired extent, all in the manner conventionally accomplished in the manufacture of aluminum complex greases. If milling is accomplished, the product from the milling zone can be moved into storage container loading, or the like, as desired. Any convenient or desired technique for processing a product grease from a cooling zone E can be utilized, as those skilled in the art will appreciate, without departing from the spirit and scope of the present invention.

Referring to Figure 2, there is seen a simplified schematic view of one embodiment of a commercial scale operation utilizing the continuous grease making process of the present invention, such operation, for convenience, being designated in its entirety herein by the numeral 10. Operation 10 is seen to employ a plurality of first make-up tanks designated herein as 11, 12, 13 and 14, respectively, such tanks 11-14 being adapted for the preparation of starting dispersions of oxyaluminum acylates in accordance with the practice of the present invention. In each make-up tank, an oxyaluminum acylate/base oil solution, for example, is made-up. Each solution is conveniently analyzed for percent aluminum metal and any adjustment in solution components necessary to achieve a standardized prechosen composition are made. After which, preferably in a sequence, the contents of each make-up tank 11-14 is charged into a feed tank 15.

Similarly, a plurality of make-up tanks 17, 18, 19 and 20 are provided in which, respectively, solutions of,

for example, stearic acid, benzoic acid and base oil are made-up. Each acid solution is tested as by an acid number titration to determine the acid content of that tank. After any adjustments for accuracy or the like are made, the contents of a respective tank 17-20 are charged into a feed tank 21. The respective contents of feed tanks 15 and 21 may be preheated, if desired, to some prechosen temperature in order to facilitate subsequent mixing. The contents of the respective tanks 15 and 21 are charged via metering pumps such as pumps 23 and 24 into mixing zone 25. The mixing zone 25 is here shown as a static mixer which is a particularly convenient and presently preferred unit for accomplishing mixing of materials from feed tanks 15 and 21. A product mixture from the mixing zone (static mixer) 25 is continuously moved into a reaction zone 26. Zone 26 is here comprised of an elongated tube 27, wherein static mixer vanes 28 are positioned. In addition, the tube 27 is provided with a heating mantle or jacket through which heating fluid (such as heated oil or a vaporized heat exchange material, such as the type available commercially under the trademark "DOWTHERM" from Dow Chemical Company, Midland, Michigan, is circulated. Thus, as a product mixture moves continuously through the tube 27, it is continuously mixed so as to prevent localized overheating or underheating of a given location from occurring. The jacket temperature on tube 27 and the length of tube 27 are so regulated and chosen that by the time material issues from the tube 27, a substantially complete reaction has taken place between the oxyaluuminum acylate material and the carboxylic acid material present, so that the product issuing from the tube 27 constitutes a product grease of this invention.

Although not shown in Figure 2, but as chemical engineers will appreciate, it may be advisable to place

a holding tank, or the like, after the reaction zone 26 in order to provide for flow variations or the like in the continuous process stream involved.

The product from the reaction zone 26 is moved by a pump 31, if desired, into a heat exchange zone 32, so as to controllably cool the product grease to some predetermined lower temperature, after which the so-cooled product grease can be admixed with additives from hold tank 33, the addtives being charged along with the so-cooled grease into a mixing zone 34, such mixing zone 34 being conveniently a static mixer or the like, as those skilled in the art will appreciate. The product from mixing zone 35, if desired, can then be directly moved twoards a storage area 35 or the like (not detailed herein, but conventional).

Alternatively, instead of the procedure above related, a product from the reaction zone 26 can be moved into a mixing zone 37 by appropriate seetings for valves 38 and 39. In mixing zones 37, a product grease can be diluted with a dilutant oil from a reservoir tank 40. To the dilutant oil 40 can be added additives, if desired, from a holding tank 41, the tank 41 being conveniently provided with a valve 42 for regulating flow of additives from tank 41 into oil from reservoir 40 in feed line 43. The flow through line 43 is conveniently regulated into the mixing zone 37 by a metering pump 44. From the mixing zone 37,the product mixed diluted grease is led through pipe 46, passed open valve 47 and into the heat exchanger 32, wherein the product mixture may be reduced to some lower prechosen temperature, which is lower than the temperature produced in the mixed diluted grease as a result of mixing with the product grease the ambient temperature dilutant oil. As those skilled in the art will appreciate, it is possible

that the product from the mixing zone 37 is cool enough for immediate discharge from the mixing zone 37 to a storage area 35. If further cooling and heat exchanger 32 is desirable or necessary, the diluted product grease from exchanger 32 can be directly diverted to the storage area 35, if desired.

If desired, a product grease produced by the process of the present invention can be subjected to a milling operation of the type conventionally utilized in the aluminum complex grease making art. For example, a product grease stream leaving mixing zone 34 can be diverted using valves 48 and 49 in proper seuqence into a milling zone 50, after which the discharge from the milling zone 50 is either moved directly to a storage area 35 or (by, for example, regulating an appropriate valve network comprised, for example, of valves such as 52 and 53) into a further mixing zone 64 wherein the milled grease can be continuously recombined with product grease which is unmilled and which is derived from the product stream produced from the mixing zone 34 (assuming an appropriate configuration for valves 48 and 54, for example). In order to obtain a proper ratio of milled grease to unmilled grease, an appropriate ratio flow control 65 could be employed, in the embodiment shown, for valves 48 and 49, as those skilled in the art will appreciate. In such a blending of milled with unmilled grease, the valves 53 and 55 would be closed, and the product from the mixing zone 64 would be moved into a storage 35.

EMBODIMENTS

The present invention is further illustrated by reference to the following Examples. Those skilled in the art will appreciate that other and further embodiments are obvious and within the spirit and scope of this invention

from the teachings of these present Examples taken with the accompanying specification.

Example 1

Preparation of Mixed Oxyaluminum Acylate

To a three neck 500 ml flask or pot is added 100 grams of a grease base oil having a viscosity at 100°F of 1766 SUS. This is a dark, amber, heavy viscous oil mixture used for grease manufacture. To this oil in such flask is added 30 grams of acetic acid, 71 grams of hydrogenated tallow fatty acids and 30.5 grams of benzoic acid and finally 102.1 grams aluminum isopropoxide. This mixture is stirred and the temperature gradually increased to a point where isopropanol begins to distill off. As the distillation continues, temperature readings are taken at 30 minute intervals and the following Table results:

Table 1

POT TEMPERATURE	VAPOR TEMPERATURE
95°C	82°C
88°C	82°C
88°C	82°C
93°C	82°C
110°C	82°C
120°C	82°C
172°C	83°C
190°C	89°C
210°C	90°C
215°C	80°C
230°C	88°C

During the distillation, a total of 2 moles of isopropyl alcohol are removed after which one mole of isopropyl acetate is removed, all on the theoretical basis.

The reaction mixture is thereafter allowed to cool to room temperature. The product assays at 5.72 percent aluminum, indicating a 51.9 percent solution of mixed oxyaluminum stearate/benzoate with a stearate/benzoate mole ratio of 1.1 in grease base oil. At room temperature, this product is a soft paste having a generally clear dark amber hue. This product is pourable at a temperature of about 50-60°C.

In place of the grease base oil, one may employ any inert solvent medium having a relatively high boiling point, such as, for example, Magie Oil No. 47 (which is an all aliphatic oil made by Magie Oil Company having an approximate boiling point of 240°C), an ester, such as one of the type used to make synthetic lubricating oils, or the like.

Example 2

Grease Preparation

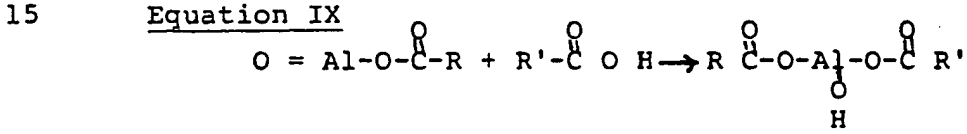
To 85 grams of the same grease base oil as employed in Example 1 contained in a beaker is added 10.2 grams of the product obtained in Example 1. The resulting mixture is stirred and gradually heated to 110°C where it is observed that a clear solution results. At this point, there is added to the heated system simultaneously 3.7 grams of hydrogenated tallow fatty acid and 1.1 gram of benzoic acid with stirring. The amount of the acids added in this grease making example is calculated in such a manner as to produce a "final soap" in situ which has a molar ratio of 1.1 to 0.9 fatty to benzoyl groups and which contains one hydroxyl group per aluminum atom. Heating is continued and the temperature is gradually raised to 140°C. After the acids are added a slight haze forms which disappears by the time the temperature reaches 140°C. With continued heating, the temperature of the system is

thereafter gradually increased to 180°C at which time heating is discontinued. After the acids are added, and as the temperature is thus gradually increased, gradual thickening of the system is observed. The thickening takes place in less than one hour due to the elimination of the step where alcohol is boiled off.

The product at room temperture is a clear grease, dark amber in color.

The thickening during heating as above described indicates the occurrence of a reaction between the oxyaluminum stearate benzoate and the added acids. It is believed thickening does not occur unless hydroxyl groups are present. Therefore, the reaction involved is representable by the following illustrative equation:

Equation IX



where R represents a mixture of fatty acid radicals and benzoic acid radicals in a molar ratio of 1:1 present in the oxyaluminum acylate starting material and R' represents a mixture of fatty acid radicals and benzoate acid radicals in a molar ratio of 1.2 to 0.8 added during grease manufacture. Thus, this grease product contains a diacyl mono hydroxy aluminum soap derived from the starting oxyaluminum stearate benzoate, from Example 1, through reaction with the added stearic and benzoic acids. Said soap containing a molar ratio of 1.1 to 0.9 fatty radicals to benzoyl radicals and having approximately one hydroxyl group per aluminum atom.

In this Example, the stearic acid could have been added before the benzoic acid, if desired. Also, in this Example, the ratio of acids added could be changed so that the final soap contains a molar ratio of 1:1 fatty radicals

to benzoyl radicals, or any other ratio desired.

In this Example, the starting materials are substantially anydrous. No water or alcohol is added to the system during processing and no water or alcohol is evolved from the system during processing. Further, no water or alcohol is found to be present in the system after processing.

Example 3

To a 500 ml three neck flask is added 76.9 g of a naphthenic base lubricating oil having a viscosity of 172 SUS (Saybolt Universal Seconds) at 100°F. To this oil is added 36.4 g hydrogenated tallow fatty acids, 31.3 grams benzoic acid, 23.1 g acetic acid, and finally 78.5 grams aluminum isopropoxide. This mixture is stirred and the temperature gradually increased to a point where isopropyl alcohol begins to distill off. As the distillation continues, temperature readings are taken at 60 minute intervals and the following Table results:

Table II

POT TEMPERATURE	VAPOR TEMPERATURE
90°C	82°C
90°C	82°C
127°C	82°C
177°C	93°C
207°C	93°C
202°C	98°C
205°C	95°C

During the distillation, a total of 2 moles of isopropyl alcohol are removed after which one mole of isopropyl acetate is removed, all on a theoretical basis. The reaction mixture is thereafter allowed to cool to room temperature. The product is a high melting resili nt solid,

dark amber and relatively clear. The aluminum was analyzed to be 6.19% which indicates a 50% solution of mixed oxy-aluminum stearate (hydrogenated tallow) benzoate in 172 SUS naphthenic base oil. The product has a stearate to benzoate mole ratio of 1:2.

Example 4

To 85 grams of the same grease base oil as employed in Examples 1 and 2 contained in a beaker is added 9.4 grams of the product from Example 3. The mixture is stirred and the temperature is raised to 170°C and held for 45 minutes until the mixed aluminum acylate dissolved. The temperature is then allowed to recede to 110°C at which point there is added to the system simultaneously 4.7 grams of hydrogenated tallow fatty acids and 0.8 grams benzoic acid with stirring. The temperature is then raised slowly to 180°C at which time the heating is discontinued. Gradual thickening is observed after the acids are added and as the temperature is gradually increased. The product is a clear heavy grease, dark amber in color. The aluminum soap which was made in situ is a mixed soap with a molar ratio of 1.1 to 0.9 fatty to benzoyl radicals and which contains one hydroxyl group per aluminum atom.

Example 5

To a 500 ml three necked flask is added 108.8 grams Magiesol 47 (Magie Bros. Oil Co., Franklin Park, IL) which is an aliphatic solvent having a boiling point of approximately 470°F. To this solvent is added 75.73 grams hydrogenated tallow fatty acids, 16.28 grams benzoic acids and 81.68 grams aluminum isopropoxide. This mixture is under agitation and the temperatures slowly increased to a point where isopropyl alcohol starts to distill off. When the pot temperature reaches 115°C and one mole of alcohol has been taken

off on a theoretical basis, 24 grams of acetic acid are introduced into the reaction mixture through a dropping funnel. Agitation is continued and the flask temperature held between 115 and 122°C until the second mole of isopropyl alcohol (theoretical basis) is distilled off. The temperature is gradually increased and isopropyl acetate begins to distill over. By the time the temperature reaches 246°C, one theoretical mole of isorpropyl acetate has been distilled off. The reaction mixture is thereafter allowed to cool to room temperature. The product is a liquid light amber in color and substantially clear. The product is analyzed to contain 4.72% aluminum which indicates that the product is 47.5% oxyaluminum stearate/benzoate in Magiesol 47 aliphatic solvent. The product has a stearate to benzoate mole ratio of 2:1 and is representative of the prior art compounds disclosed in Harson British Patent No. 825,878.

Example 6

283.5 grams of the same base oil as employed in Examples 1, 2 and 4, contained in a beaker is added 12.4 grams of th product from Example 5. The mixture is heated with stirring to a temperature of 130°C at which temperature there is added to the system simultaneously 2.65 grams of hydrogenated tallow fatty acid and 1.5 grams of benzoic acid with stirring. As both acids are simultaneously added, it is observed that the resultant mixture of lubricating base oil, aluminum derivative and acids turns cloudy. The temperature is then raised to 200°C for 15 minutes. During the time of increasing the temperature the cloud diminishes somewhat, but does not entirely disappear. Holding the mixture at a temperature of 200°C for 15 minutes

does not improve the clarity of the mixture. Although thickening occurs during the increasing of the temperature, the viscosity of the mixture is not as high as greases made with compounds of the present invention.

Example 7

To a 500 ml three neck flask is added 78.1 grams isopropyl octoate. To this high boiling ester is added 51.1 grams isostearic acid (Emersol 875, Emery Industires, Inc.), 21.9 grams benzoic acid, 21.6 grams acetic acid and finally 73.5 grams aluminum isopropoxide. This mixture is stirred and the temperature gradually increased to a point where isopropyl alcohol begins to distill off. As the distillation continues, temperature readings are taken at bi-hourly intervals and the following Table results.

Table III

POT TEMPERATURE	VAPOR TEMPERATURE
97°C	82°C
125°C	85°C
156°C	88°C
176°C	105°C
180°C	100°C
180°C	98°C
185°C	93°C
194°C	98°C
201°C	104°C

During the distillation, a total of 2 moles of isopropyl alcohol are removed after which one mole of isopropyl acetate is removed, all on a theorectical basis. The reaction mixture is thereafter allowed to cool to room temperature.

The product is a light amber liquid relatively clear. The aluminum is analyzed to be 5.70% which indicates a 50% solution of oxyaluminum isostearate/benzoate in isopropyl octoate. The product has an isostearate to benzoate mole ratio of 1:1, and can be converted to the solvent free state by distilling off the isopropyl octoate preferably under reduced pressure.

Example 8

To 84.5 grams of the same grease base oil as employed in Example 1 contained in a beaker is added 10.8 grams of the product solution obtained in Example 7. The resulting clear mixture is stirred and gradually heated to 90°C. At this point, there is added to the heated system simultaneously 3.2 grams hydrogenated tallow fatty acids and 1.4 grams benzoic acid with stirring. The amount of the acids added in this example is calculated in such a manner as to produce a "final soap" in situ which has a molar ratio of 1:1 fatty to benzoyl groups and which contains one hydroxyl group per aluminum atom and can be described as a balanced mixed soap. The temperature is then raised slowly to 190°C at which time the heating is discontinued. Gradual thickening is observed after the acids are added and as the temperature is increased. The product is a clear, heavy grease, dark amber in color.

Example 9

To 84.5 grams of the same grease base oil as employed in Example 1 contained in a beaker is added 10.4 grams of the product solution obtained in Example 7. The resulting clear mixture is stirred and gradually heated to 90°C. At this point, there is added to the heated system simultaneously 3.7 grams hydrogenated tallow fatty acids and 1.1 grams benzoic acid with stirring. The temperature is then raised slowly to

190°C at which time the heating is discontinued. Gradual thickening is observed after the acids are added and as the temperature is increased. The product is a clear, heavy grease, dark amber in color. The aluminum soap which is made in situ in this example is a mixed soap with a slightly unbalanced molar ratio of fatty to benzoyl groups of 1.1 to .9 and contains one hydroxyl group per aluminum atom.

Example 10

To a 3 litre glass resin kettle equipped with a stirring motor and a lid with 3 openings is added the following ingredients: 687.3 grams hydrogenated tallow fatty acids, 503.7 grams benzoic acid, and 673.9 grams isopropyl alcohol. This mixture is heated until it becomes a homogenous clear solution at approximately 60°C. To this mixture is then added 673.9 grams aluminum isopropylate. Heat is then applied to the reaction vessel until it rises to a temperature where isopropyl alcohol begins to distill off. The distillation is continued and periodic temperature readings are taken and the following Table results:

TABLE I

Time	Pot Temperature	Vapor Temperature
0	85°C	81°C
1 1/2 hrs.	85°C	81°C
2 hrs	85°C	80°C
3 3/4 hrs.	205°C	65°C

During the above distillation procedure, both the added isopropyl alcohol and the isopropyl alcohol produced by the reaction process are removed from the reaction vessel on a theoretical basis. Heating is continued at 200°C for 1 1/2 hours and then the reaction mixture is allowed to cool. The product is a light amber clear oily liquid. The aluminum is analyzed to be 5.86% and by further analysis it is determined that the oxyaluminum acylate contained 85% benzoic radicals.

The oxyaluminum acylate is dissolved in a mixture of isopropyl benzoate and isopropyl hydrogenated tallowate.

Example 11

To 303.7 grams of a grease base oil having the viscosity at 100°F of 1766 SUS is added 29.8 grams of the compound from Example 10. The resulting mixture is stirred and gradually heated to 90°C where it is observed that a homogenous, relatively clear mixture results. At this point, there is added to the heated system simultaneously 16.3 grams hydrogenated tallow fatty acid and .1 grams benzoic acid with stirring. The amount of ingredients added to the base oil in this grease making Example is calculated in such a manner as to produce a final grease with an aluminum content of 0.5% aluminum metal, a fatty to benzoic ratio of 1.1 to .9 and a ratio of 1.92 moles total acids per atoms of aluminum. Heating is continued and the temperature is gradually raised to a temperature of 200°C and the mixture is held at 200°C for one-half hour. After the acids are added, and as the temperature is thus gradually increased, gradual thickening of the system is observed. The reaction mixture remains clear throughout the heating process and results in a clear grease. No acetic acid odor is detected during this process. After the reaction mixture is held for one-half hour at 200°C, it is allowed to cool and physical properties are determined. The resultant clear grease has a dropping point of 468°F and an unworked penetration of 307. After working 60 strokes in a standard grease worker, the penetration is 314. The grease remains soft and pliable after standing overnight indicating the absence of false set properties.

Example 12

Attempt to Make 85 Mole % Benzoic Mixed Oxyaluminum Acylate Via Acetic Acid Method

To a 1000 ml. 3-neck flask is added the following ingredients: 183.2 grams Coray 22 which is lubricating base

oil having an approximate viscosity of 100 SUS at 100°F, 50 grams isopropyl alcohol, 48 grams glacial acetic acid, 33.3 grams hydrogenated tallow fatty acids, and 83.0 grams benzoic acid. The temperature of this mixture is raised to 55°C at which point 163.4 grams powdered aluminum isopropylate is added to the flask. This mixture is stirred and gradually increased to a point where ispropanol begins to distill off. As the distillation continues, periodic temperature readings are taken and the following Table results:

TABLE II

Hours	Pot Temperature	Vapor Temperature
0	89°C	81°C
0.5	96°C	81°C
1.0	156°C	80°C
1.25	198°C	81°C
1.5	202°C	65°C
2.0	210°C	30°C
2.5	194°C	30°C
4.0	192°C	30°C

During this distillation procedure, the material in the flask never goes through a clear state and does not end up clear. It is only thin when the temperature rises to approximately 200°C; however, it is still an opaque liquid at this point. Very little distillate is taken off between the temperatures of 156°C and 200°C indicating that only a small amount of ispropyl acetate ester is formed by this reaction. The total distillate measures 149 grams and accounts for little more than the theoretical ispropanol released by the reaction with the acids plus the 50 grams ispropyl alcohol added to facilitate dispersion of the initial materials.

Example 13

The procedure in Example 12 is repeated to make sure that the results are reliable. The same quantities of ingredients are added in the same order and the mixture is stirred and the temperature gradually increased to a point where isopropanol begins to distill off. As the distillation continues temperature readings are taken and the following Table results:

TABLE III

Hours	Pot Temperature	Vapor Temperatur
0	90°C	81°C
.5	93°C	81°C
1.0	180°C	81°C
1.5	200°C	35°C
2.0	180°C	23°C
2.5	202°C	23°C
3.5	196°C	23°C
4.0	194°C	23°C
4.5	197°C	23°C
5.0	197°C	23°C
5.5	203°C	23°C
6.0	200°C	23°C

During this procedure extra care is taken to ensure that excessive heat does not damage or interfere with the reaction. At no time does the temperature ever exceed 205°C. As can be seen by the Table, no appreciable isopropyl acetate is taken off as indicated by the low vapor temperatures which are recorded as the pot temperature moves towards 200°C. As with Example 3, the mixture in the flask never turns clear and remains an opaque hetergenous mixture.

Example 14

An attempt is made to make a grease from the material produced in Example 13. 302.2 grams of the same grease base oil as employed in Example 11 are placed in a beaker which contains a magnetic stir bar. This beaker is placed on a hot plate and heated with stirring to a temperature of 160°C. 31.4 grams of the product obtained from Example 13 is melted and added to the oil. During this addition, it is noted that solid particles start forming immediately. The size of the particles are about 1/16th of an inch to 1/8th of an inch in diameter. The mixture is then heated to 200°C in an attempt to disperse the particles, but this is not successful. The temperature is then lowered to 95°C at which temperature 16.3 grams hydrogenated tallow fatty acid are added slowly to the mixture under agitation. Then, .1 gram benzoic acid is added immediately following this addition of the hydrogenate tallow fatty acids and it is noted that the temperature is 100°C when the benzoic acid is in. The particles noted above do not disappear. The mixture is then raised again to a temperature of 200°C. The mixture thickens slightly, but still contains the solid opaque particles. This is not usable grease.

Example 15

Preparation of Mixed Osyaluminum Acylate Containing Benzoic Acid Via the Acetic Acid Method

To a 1000 ml. 3-neck flask is added 198.4 grams Coray 22 oil (lubricating base oil having an approximate viscosity of 100 SUS at 100°F). To this oil in such a flask is added the following ingredients: 50 grams isopropyl alcohol, 48 grams acetic acid, 55.5 grams hyrogenated tallow fatty acids, 73.3 grams benzoic acid. This mixture is warmed slightly to produce a homogenous clear liquid. The temperature is then raised to 65°C and at this point is added to the system 163.4 grams aluminum isopropylate.

This mixture is stirred and the temperature gradually increased to a point where isopropanol begins to distill off. As the distillation continues periodic temperature readings are taken and the following Table results:

TABLE IV

Time	Pot Temperature	Vapor Temperature
0	88°C	81°C
.5	88°C	81°C
1.0	90°C	81°C
1.5	99°C	81°C
2.0	110°C	81°C
2.5	180°C	84°C
3.0	200°C	87°C
3.5	200°C	87°C
4.0	204°C	87°C

During the distillation a total of 2 moles ispropyl alcohol are removed after which 1 mole of isopropyl acetate is removed all on a theoretical basis. The reaction mixture is thereafter allowed to cool to room temperature. The product is a clear solid amber material having a melting point of approximately 130°C and by calculation is found to contain 5.44% aluminum indicating a 40.9% solution (in 100 SUS 100°F lubricating oil) of mixed oxyaluminum stearate/benzoate wherein the mole percent benzoate is 75%.

Example 16

Grease Made From Example 15

303.5 grams of the same grease base oil as employed in Example 11 are placed in a beaker which contains a magnetic stir bar. This beaker is placed on a hot plate and is heated with stirring to a temperature of 160°C. 32.2 grams of the product obtained from Example 15 is melted and added to the oil. Hearing is continued with stirring and the temperature

is gradually raised to a temperature of 200°C and it is observed that the mixture is uniformly dispersed. The temperature is then lowered to 95°C at which temperature 14.5 grams hydrogenated tallow fatty acid are added slowly to the mixture under agitation. Then, 0.9 grams benzoic acid are added immediately following this addition of the hydrogenated tallow fatty acids. The amount of ingredients added to the base oil in this grease making Example is calculated in such a manner as to produce a final grease with an aluminum content of 0.5% aluminum metal, a fatty to benzoic ratio of 1.1 to .9 and a ratio of 1.92 moles total acids per atom of aluminum. The mixture is again raised to a temperature of 200°C and during such period of heating is observed an acid odor resembling acetic acid.

After the acids are added, and as the temperature is thus gradually increased, gradual thickening of the system is observed. The reaction mixture remains clear throughout the heating process and results in a clear grease. After the mixture is held for 15 minutes at 200°C, it is allowed to cool and physical properties are determined. The resultant clear grease has a dropping point of 479°F and an unworked penetration of 301. After working 60 strokes in a standard grease worker, the penetration is 314.

Example 17

To a 22 litre 3-neck flask is added the following ingredients: 4,998.6 grams hydrogenated tallow fatty acid, 2,197.8 grams benzoic acid and 3,816 grams isopropanol. This mixture is heated to approximately 60°C at which point the mixture is a clear low viscosity homogenous liquid. To this mixture is then added 3,675.6 grams granulated aluminum isopropylate. Heat is applied to the flask and the temperature gradually raised to the point where isopropanol begins to distill off. As the distillation continues temperature readings are taken at 60 minute intervals

and the following Table results:

TABLE V

Pot Temperature	Vapor Temperature
84°C	81°C
85°C	81°C
85°C	81°C
85°C	81°C
93°C	81°C
100°C	81°C
120°C	81°C
166°C	84°C

At this point, the heating causes the temperature to begin rising much more rapidly and it reaches 200°C within another hour. During the time of the first and second steps of heating, both the added ispropyl alcohol and 2 moles of produced isopropyl aocohol on a theoretical basis are removed from the flask. The temperature is then maintained at 200°C for one more hour after which it is allowed to cool. The product is a light amber clear liquid which is analyzed to be 5.67% aluminum and by further analysis it is determined that the oxyaluminum acylate so produced contained 75.3% benzoic radicals. The oxyaluminum acylate so produced contained 75.3% benzoic radicals. The oxyaluminum acylate is dissolved in a mixture of isopropyl benzoate and isopropyl hydrogenated tallowate.

Example 18

Grease From Example 17

To 303.8 grams of a grease base oil having the viscosity at 100°F of 1766 SUS is added 30.8 grams of the compound from Example 17. The resulting mixture is stirred

and gradually heated to 90°C where it is observed that a clear solution results. At this point there is added to the heated system simultaneously 14.3 grams hydrogenated tallow fatty acid and 0.9 grams benzoic acid with stirring. The amount of ingredients added to the base oil in this grease making Example is calculated in such a manner as to produce a final grease with an aluminum content of 0.5% aluminum metal, a fatty to benzoic ratio of 1.1 to 0.9 and a ratio of 1.92 moles total acids per atom of aluminum. Heating is continued and the temperature is gradually raised to a temperature of 200°C and the mixture is held at 200°C for one-half hour. After the acids are added, and as the temperature is thus gradually increased, gradual thickening of the system is observed. The reaction mixture remains clear throughout the heating process and results in a clear grease. No acetic acid odor is detected during this process. After the reaction mixture is held for one-half hour at 200°C, it is allowed to cool and physical properties are determined. The resultant clear grease has a dropping point of 509°F and an unworked penetration of 245. After working 60 strokes in a standard grease worker, the penetration is 286. The grease remains soft and pliable after standing overnight, indicating the absence of false set properties.

Example 19

To a 1,000 ml. 3-neck flask the following ingredients are placed: 333.2 grams hydrogenated tallow fatty acid, 48.8 grams benzoic acid, and 67.5 grams isopropyl alcohol. This mixture is heated to approximately 80°C at point the contents of the flask comprises a homogenous clear liquid system. To the flask next is added 272.3 grams of a 60% solution of aluminum isopropylate in isopropanol. This mixture is stirred and heat is applied to the flask until isopropyl alcohol begins to distill off. As the

distillation continues, temperature readings are taken at 30 minute intervals and the following Table results:

TABLE I

Pot Temperature	Vapor Temperature
87°C	80°C
96°C	80°C
114°C	80°C
120°C	80°C
130°C	80°C
155°C	80°C

At this point, 92% of the theoretical isopropyl alcohol has been distilled off. More heat is then applied to the flask causing the temperature to rise to 200°C over a period of an hour. During this time, the remainder of the alcohol on a theoretical basis is distilled off of the reaction mixture. As the temperature approaches 200°, it is observed that the mixture in the flask is a low viscosity clear amber liquid. The temperature is maintained at 200°C for one hour. The reaction mixture is thereafter allowed to cool to room temperature. The product is a low viscosity clear amber liquid. The aluminum is analyzed to be 4.80% and by analysis the oxyaluminum acylate is found to have 42.7% benzoic radicals. The oxyaluminum acylate is dissolved in a mixture of isopropyl benzoate and isopropyl hydrogenated tallowate.

Example 20

Grease from Example 19

To 301.4 grams of a grease base oil having the viscosity at 100°F of 1766 SUS is added 36.4 grams of the compound from Example 19. The resulting mixture is stirred and gradually heated to 90°C where it is observed that a

clear solution results. At this point there is added to the heated system simultaneously 8.7 grams hydrogenated tallow fatty acid and 3.4 grams benzoic acid with stirring. The amount of ingredients added to the base oil in this grease making Example is calculated in such a manner as to produce a final grease with an aluminum content of 0.5% aluminum metal, a fatty to benzoid ratio of 1.1 to 0.9 and a ratio of 1.92 moles total acids per atom of aluminum. Heating is continued and the temperature is gradually raised to a temperature of 200°C and the mixture is held at 200°C for one-half hour. After the acids are added, and as the temperature is thus gradually increased, gradual thickening of the system is observed. The reaction mixture remains clear throught the heating process and results in a clear grease. No acetic acid odor is detected during this process. After the reaction mixture is held for one-half hour at 200°C, it is allowed to cool and physical properties are determined. The resultant clear grease has a dropping point of 495° and an unworked penetratio of 311. After working 60 strokes in a standard grease worker, the penetration is 288. The grease remains soft and pliable after standing overnight indicating the absence of false set properties.

Example A

Starting Mixture for Continuous Process

A carboxylic acid mixture suitable for use in the continuous process of the present invention is prepared as follows:

Into a 15 gallon expoxy lined drum is charged 50.6 pounds of a grease base oil having a viscosity at 100°F of no more than about 1800 SUS. This drum is provided with an impeller driven by an electric motor and also with an external electric heating device to heat the contents to a predetermined temperature. The oil is agitated and raised

to a temperature of about 90°C after which is added 4.75 pounds of hydrogenated tallow fatty acid and 0.25 pounds benzoic acid.

Example B

An oxyaluminum acylate composition suitable for use in the continuous process of the present invention is prepared as follows:

To a 22 litre 3-neck flask is added the following ingredients: 4,499 grams hydrogenated tallow fatty acid, 2,418 grams benzoic acid and 3,672 grams isopropanol. This mixture is heated to approximately 60°C at which point the mixture is a clear low viscosity homogenous liquid. To this mixture is then added 3,675.6 grams granulated aluminum isopropylate. Heat is applied to the flask and temperature gradually raised to the point where isopropanol begins to distill off. As the distillation continues periodic temperature readings are taken and the following Table results:

TABLE II

Hours	Pot Temperature	Vapor Temperature
0	84°C	81°C
2	84°C	81°C
2.5	91°C	81°C
3	95°C	81°C
6.5	100°C	80°C
7.5	170°C	80°C
8	201°C	33°C
9.5	207°C	33°C

During the heating, both the added isopropyl alcohol and 2 moles of produced isopropyl alcohol on a theoretical basis are removed fromthe flask. The flask is then allowed to cool. The product is a light amber clear liquid which is

analyzed to be 5.96 aluminum and by further analysis it is determined that the oxyaluminum acylate so produced contained 81% benzoic radicals. The oxyaluminum acylate is dissolved in a mixture of isopropyl benzoate and isopropyl hydrogenated tallowate.

Example C

A carboxylic acid mixture suitable for use in the practice of the present invention is prepared as follows:

To a similarly equipped drum as used in Example A is charged 50.75 pounds of the same grease base oil as identified in Example A. The oil is agitated and heated to a temperature of about 90°C after which is added 5.2 pounds of hydrogenated tallow fatty acids and 0.15 pounds benzoic acid.

Example D

A carboxylic acid mixture suitable for use in the practice of the present invention is prepared as follows:

To a similarly equipped drum as used in Examples A and C is charged 50.75 pounds of the same grease base oil as used in Example A. The oil is agitated and heated to a temperature of about 90°C after which is added 6.0 pounds of hydrogenated tallow fatty acids.

Example E

An oxyaluminum acylate composition suitable for use in the practice of the present invention is prepared as follows:

To a 22 litre 3-neck flask is added 2,332 grams Coray 22 oil (lubricating base oil having an approximate viscosity of 100 SUS at 100°F). To this oil in such a is added the following ingredients: 1,320 grams acetic acid, 2,996 grams hydrogenated tallow fatty acids, 1,342 grams

benzoic acid. This mixture is warmed slightly to produce a homogenous clear liquid. The temperature is then raised to 65°C and at this point is added to the system 4,492 grams aluminum isopropylate. This mixture is stirred and the temperature gradually increased to a point where isopropanol begins to distill off. As the distillation continues periodic temperature readings are taken and the following Table results:

TABLE III

Time	Pot Temperature	Vapor Temperature
0	90°C	82°C
1.0	90°C	82°C
1.5	92°C	82°C
2.5	97°C	82°C
3.0	106°C	82°C
4.0	135°C	84°C
4.5	160°C	96°C
5.5	185°C	108°C
6.0	195°C	94°C
6.5	210°C	115°C
7.0	215°C	115°C
7.5	218°C	112°C

After seven and one-half hours of total elapsed time, the heat is turned off and an additional 2,332 grams Coray 22 base oil were added to the flask. During the distillation a total of 2 moles isopropyl alcohol are removed after which 1 mole of isopropyl acetate is removed all on a theoretical basis. The reaction mixture is thereafter allowed to cool to room temperature. The product is a clear high viscosity amber liquid which is found by analysis to contain 5.77% aluminum indicating approximately 50% solution (in 100 SUS 100°F lubricating oil) of mixed oxyaluminum stearate/benzoate wherein the mole percent benzoate is 50%.

Example 21

Continuous Process

Into a 15 gallon epoxy lined drum is charged 50.6 pounds of a grease base oil having a viscosity at 100°F of no more than 1800 SUS. This drum is provided with an impeller driven by an electric motor and also with an external electric heating device to heat the contents to a predetermined temperature. The oil is agitated and raised to a temperature of about 90°C after which is added thereto about 10.25 pounds of an oxyaluminum acylate intermediate composition prepared as described in Example 17 (above). The entire resulting mixture is brought to a temperature estimated to be about 90°C, and mixing is continued until a homogenous clear solution is obtained, wherein the total weight percentage of mixed oxyaluminum acylate is estimated to be about 7.0 weight percent. The total weight percent of high boiling isopropanol mixed acid esters is estimated to be about 9.7 weight percent.

Example G

A carboxylic acid mixture suitable for use in the practice of the present invention as prepared as follows:

To a similarly equipped drum as used in Example B, is charged 51.2 pounds of the same grease base oil as is identified in Example B. The oil is agitated and heated to a temperature of about 90°C after which is added 3.3 pounds of hydrogenated tallow fatty acids and 0.9 pounds benzoic acid.

The drum containing the oxyaluminum acylate solution described above and the drum containing the acid solution as described in Example A are both connected by suitable piping to a Kenix standard static mixer, (which has an internal diameter of 3/8" and contains 21 elements) through metering pumps. The static mixer is in turn interconnected through a surge tank/booster pump combination device with

a coiled copper tube, approximately 8 feet in length and having an internal diameter of about 3/8 inches. This coiled tubing is immersed in a drum of heat exchange oil which has been heated to a temperature sufficient to maintain the interior of the coiled tubing at a temperature of not less than 200°C. The discharge end of the reactor coil is placed over a 15 gallon receiving drum which is equipped with a cooling coil through which water is circulated.

The contents of the first drum are charged into the feed end of the static mixer at a rate estimated to be about 0.125 gallons per minute. Thus, the total amount of aluminum in the combined mixture entering the static mixer is calculated to be about 0.5 weight percent (based upon total composition in the static mixer). Also, in the static mixer, the mole ratio of the total amount of aluminum to total carboxylic acid material is calculated to be about 1:1.92, the total carboxylic acid material here defined as the acids added from the second rum combined with the acylate groups present as part of the oxyaluminum acylate composition. The effluent fromt he static mixture is believed to be homogenous and uniform in composition. After passing through the surge tank/booster pump device, the flow rate of the combined mixture entering the tube reactor is estimated to be about 0.25 gallons/min. The temperature of the material being discharged from the reactor is estimated to be about 200°C. The product from the tubular reactor is discharged into the receiving container and cooled to a lower temperature by the cooling coils. The product is a clear grease having a dropping point estimated to be higher than 475°F and an unworked penetration estimated to be less than 300.

Example 22

Into a 15 gallon epoxy lined drum is charged 50.75 pounds of a grease base oil having a viscosity of 100°F of no more than 1800 SUS. This drum is provided with an impeller driven by an electric motor and also with an external electric heating device to heat the contents to a predetermined temperature. The oil is agitated and raised to a temperature of about 90°C after which is added 9.8 pounds of an oxyaluminum acylate intermediate composition prepared as described in Example B (above). The entire resulting mixture is brought to a temperature estimated to be about 90°C, and mixing is continued until a homogenous clear solution is obtained, wherein the total weight percentage of mixed oxyaluminum acylate is estimated to be about 6.7 weight percent, the total weight percent of high boiling isopropanol mixed acid esters is estimated to be about 9.5 weight percent, with the balance up to 100 weight percent being the grease base oil as described above.

The drum containing the oxyaluminum acylate solution described above and the drum containing the acid solution as described in Example C are both fed into the same equipment as described in Example 21.

The contents of the first drum are charged into the feed end of the static mixer at a rate estimated to be about 0.125 gallons per minute while the contents of the second drum are similarly charged at a rate estimated to be about .125 gallons per minute. Thus, the total amount of aluminum in the combined mixture entering the static mixer is calculated to be about 0.5 weight percent (based upon total composition in the static mixer). Also, in the static mixer, the mole ratio of the total amount of aluminum to total carboxylic acid material is calculated to be about 1:1.92, the total carboxylic acid material here defined as the acids added fromt he second drum combined with the

acylate groups present as part of the oxyaluminum acylate composition. The effluent from the static mixture is believed to be homogenous and uniform in composition. After passing through the surge tank/booster pump device, the flow rate of the combined mixture entering the tube reactor is estimated to be about 0.25 gallons/min. The temperature of the material being discharged from the reactor is estimated to be about 200°C. The product from the tubular reactor is discharged into the receiving container and cooled to a lower temperature by the cooling coils. The product is a clear grease having a dropping point estimated to be higher than 475°F and an unworked penetration estimated to be less than 300.

Example 23

Into a 15 gallon epoxy lined drum is charged 50.75 pounds of a grease base oil having a viscosity at 100°F of no more than 1800 SUS. This drum is provided with an impeller driven by an electric motor and also with an external electric heating device to heat the contents to a predetermined temperature. The oil is agitated and raised to a temperature of about 90°C after which is added 9.8 pounds of an oxyaluminum acylate intermediate composition prepared as described in Example B (Above). The entire resulting mixture is brought to a temperature estimated to be about 90°C, and mixing is continued until homogenous clear solution is obtained, wherein the total weight percentage of mixed osyaluminum acylate is estimated to be about 6.7 weight percent, the total weight percent of high boiling isopropanol mixed acid esters is estimated to be about 9.5 weight percent, the balance up to 100 weight percent being the grease base oil as described above.

The drum containing the oxyaluminum acylate solution described above and the drum containing the acid solution as described in Example D are both fed into the same equip-

ment as described in Example 21.

The contents of the first drum are charged into the feed end of the static mixer at a rate estimated to be about 0.125 gallons per minute while the contents of the second drum are similarly charged at a rate estimated to be about 0.125 gallons per minute. Thus, the total amount of aluminum in the combined mixture entering the static mixer is calculated to be about 0.5 weight percent (based upon total composition in the static mixer). Also, in the static mixer, the mole ratio of the total amount of aluminum to total carboxylic acid material is calculated to be about 1:2, the total carboxylic acid material here defined as the acids added from the second drum combined with the acylate groups present as part of the oxyaluminum acylate composition. The effluent from the static mixture is believed to be homogenous and uniform in composition. After passing through the surge tank/booster pump device, the flow rate of the combined mixture entering the tube reactor is estimated to be about 0.25 gallons per minute. The temperature of the material being discharged from the reactor is estimated to be about 200°C. The product from the tubular reactor is discharged into the receiving container and cooled to a lower temperature by the cooling coils. The product is a clear grease having a dropping point estimated to be higher than 475°F, and an unworked penetration estimated to be less that 300.

Example 24

Into a 15 gallon epoxy lined drum is charged 51.2 pounds of a grease base oil having a viscosity at 100°F of no more than 1800 SUS. This drum is provided with an impeller driven by an electric motor and also with an external electric heating device to heat the contents to a predetermined temperature. The oil is agitated and raised to a temperature of about 90°C after which is added 10.1 pounds of an oxyaluminum

acylate intermediate composition prepared as described in Example E (above). The entire resulting mixture is brought to a temperature estimated to be about 90°C, and mixing is continued until a homogenous clear solution is obtained, wherein the total weight percentage of mixed oxyaluminum acylate is estimated to be about 8.2 weight percent, with the balance up to 100 weight percent being grease base oil.

The drum containing the oxyaluminum acylate solution described above and the drum containing the acid solution as described in Example F are both connected tothe same equipment as described in Example 21.

The contents of the first drum are charged into the feed end of the static mixer at a rate estimated to be about 0.125 gallons per minute while the contents of the second drum are similarly charged at a rate estimated to be about 0.125 gallons per minute. Thus, the total amount of aluminum in the combined mixture entering the static mixer is calculated to be about 0.5 weight percent (based upon total composition in the static mixer). Also, in the static mixer, the mole ratio of the total amount of aluminum to total carboxylic acid material is calculated to be about 1:1.92, the total carboxylic acid material here defined as the acids added from the second drum combined with the acylate groups present as part of the oxyaluminum acylate composition. The effluent from the static mixture is believed to be homogenous and uniform in composition. After passing through the surge tank/booster pump device, the flow rate of the combined mixture entering the tube reactor is estimated to be about 0.25 gallons per minute. The temperature of the material being discharged from the reactor is estimated to be about 200°C. The product from the tubular reactor is discharged into the receiving container and

cooled to a lower temperature by the cooling coils. The product is a clear grease having a dropping point estimated to be higher than 475°F, and an unworked penetration estimated to be less than 300.

As those skilled in the art will appreciate, minor amounts of various carboxylic acids known to the art of grease making can be present, if desired, in the reactants employed to make a grease as described herein.

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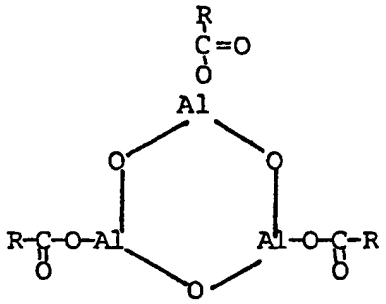
CLAIMS

The claims are:

1. Compounds of the formula $O{=}Al{-}O{-}\overset{\overset{O}{\|}}{C}{-}R$

and of the formula



wherein R is selected from the group of radicals consisting of: Type (A) aliphatic radicals each containing from 10 to 38 carbon atoms, and Type (B) aromatic radicals each containing from 6 to 16 carbon atoms, and wherein, in any given group of such compounds, the ratio of the number of radicals of said Type (B) to said Type (A) ranges from 2:3 to 19:1.

2. Compounds of claim 1 wherein said Type (A) radicals are derived from hydrogenated tallow acids and said Type (B) radicals are derived from benzoic acid.

3. Compounds of claim 1 wherein Type (A) radicals are derived from hydrogenated fish oil acids and said Type (B) radicals are derived from benzoic acid.

4. Compounds of claim 1 wherein said Type (A) radicals are derived from stearic acid and said Type (B) radicals are derived from benzoic acid.

5. Compounds of claim 1 wherein said Type (A) radicals are derived from isostearic acid and said Type (B) radicals are derived from benzoic acid.

6. A composition for use in grease manufacture comprising on a 100 weight percent total weight basis (a) from about 30 to 70 weight percent of at least one group of

compounds of claim 1, and, correspondingly, (b) from about 70 to 30 weight percent of a petroleum derived hydrocarbon having a viscosity at 100°F ranging from about 35 to 50,000 SUS, said component (a) being uniformly dispersed in said component (b).

7. In an improved process for making a grease from a starting oil having a viscosity at 110°F of from about 35 to 50,000 SUS by contacting in said oil at least one carboxylic acid material with starting organoaluminum compound, both said carboxylic acid material and said organoaluminum compound being dispersed in said oil, said contacting being carried out at a temperature sufficient to produce reaction between said carboxylic acid material and said organoaluminum compound, said contacting being continued until at least some of said organoaluminum compound has been converted into an aluminum soap and a grease is formed, the improvement which comprises employing as said starting organoaluminum compound at least one compound of claim 1, and wherein said aluminum soap is an aluminum monohydroxy diacylate.

8. A grease prepared by the process of claim 7.

9. A process for making a composition comprised of mixed oxyaluminum acylate and mixed esters of carboxylic acid material, said process comprising the steps of sequentially, first heating a mixture of an aluminum alkoxide material and a carboxylic acid material to a first temperature which is above the melting point of said carboxylic acid material but which is below the boiling point of an alkanol material while agitating said mixture, said first heating being continued for a time sufficient to form a substantially homogenous mixture, said aluminum alkoxide material being characterized by the formula: $Al(OR")_3$ where R is a lower alkyl radical, said carboxylic acid

material being comprised of at least one saturated aliphatic monocarboxylic acid containing from 8 to 40 carbon atoms per molecule, and at least one aromatic monocarboxylic acid containing from 7 to 14 carbon atoms per molecule, the mole ratio of said aromatic acid to said aliphatic acid ranging from about 2:3 to 19:1, said alkanol material being characterized by the formula: R O H, where R is defined above and where R is derived from reaction between said aluminum alkoxide material and said carboxylic acid material, the mole ratio of said aluminum alkoxide material to said carboxylic acid material ranging from about 1 to 2, secondly, heating said liquid to second temperatures in the range where said alkanol material is distilled off and maintaining said second heating for a time sufficient to remove the equivalent of at least about 2 theoretical moles of said alkanol based upon said aluminum alkoxide material, and thirdly, heating the resulting product from said second heating to gradually increasing third temperatures from a temperature corresponding to the final temperature of said second heating up to about 200°C, thereby to convert said resulting product into a homogenous final liquid having a viscosity substantially less than that of said resulting product from said second heating.

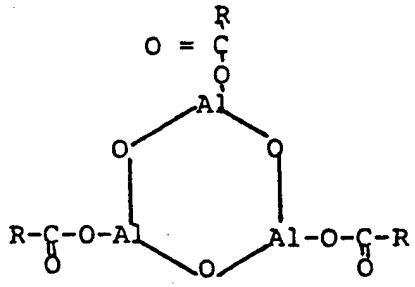
10. The process of claim 9 wherein said third heating is continued at said 200°C for a time of at least about 1/4 hour.

11. The process of claim 9 wherein, before said first heating, said carboxylic acid material is heated to said first temperature and said aluminum alkoxide material is then admixed therewith.

12. The process of claim 9 wherein up to about 50 weight percent of added alkanol material is admixed with said mixture in said first heating.

13. The process of claim 9 wherein after said third heating said final liquid is subjected to vacuum distillation so as to remove from said final liquid any organic material therein which at atmospheric temperature and pressure boils at a temperature not more than about 250°C.

14. A continuous process for making an aluminum complex grease comprising the steps of simultaneously but sequentially: mixing a first dispersion of oxyaluminum acylate material with a second dispersion of carboxylic acid material in a first mixing zone to produce a substantially homogenous product mixture, said oxyaluminum acylate material being characterized by the formula: $O{=}Al{-}O{-}\overset{O}{\overset{\|}{C}}{-}R$ and by the formula

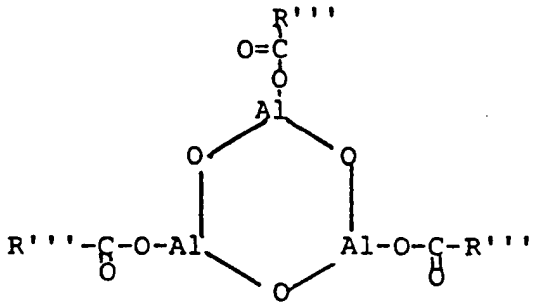


wherein R is selected from the group of radicals consisting of: Type (A) aliphatic radicals each containing from 10 to 38 carbon atoms, and Type (B) aromatic radicals each containing from 6 to 16 carbon atoms, and wherein, in any given such oxyaluminum acylate material, the mole percent of the number of radicals of said Type (B) radicals ranges from about 40 to 95 with the balance up to 100 mole percent being said Type (A) radicals, said carboxylic acid material being characterized by the formula $R'{-}\overset{O}{\overset{\|}{C}}{-}OH$ where R' is selected from the group consisting of aliphatic radicals containing from 9 through 40 carbon atoms each and aromatic

radicals containing from 6 through 28 carbon atoms each, and wherein in any given such carboxylic acid material the weight percent of said aromatic radicals ranges from and including 0 to about 70% with the balance up to 100 weight percent of all said radicals being said aliphatic radicals, the carrier liquid for each of said first and said second dispersions comprising an inert organic liquid having a viscosity at 100°F of from about 35 to 50,000 SUS, the total amount of aluminum in said product mixture ranging from about 0.01 to 2.0 weight percent based on total mixture weight, and the mole ratio of said total amount of aluminum to added said carboxylic acid material ranging from about 0.75:1.25 to 1.25:0.75, charging said product mixture into an elongated reaction zone, passing said product mixture through said reaction zone while maintaining said reaction zone at a temperature ranging from about 135 to 250°C, the residency time for said product mixture in said reaction zone being at least such that said oxyaluminum acylate susbstantially completely reacts with said carboxylic acid, thereby to form a product aluminum complex grease, removing said product grease from said reaction zone, and cooling said product grease.

15. The process of claim 14 wherein said first dispersion additionally contains an ester material having the formula $R\overset{O}{\overset{\|}{C}}-OR''$ wherein R is as defined in claim 1 and R" comprises a lower alkyl radical, and wherein, in said first dispersion, the mole ratio of said ester material to said oxyaluminum acylate material ranges from about 1.5:1 to 0.

16. The process of claim 14 wherein said first dispersion additionally contains a second oxyaluminum acylate material which is characterized by the formula $O=Al-O\overset{O}{\overset{\|}{C}}-R'''$ and by the formula



wherein R''' is as defined above but additionally includes a further group of radicals consisting of Type (C) lower alkyl radicals, and wherein, in any given such oxyaluminum acylate material on a 100 mole percent total basis, the mole percent of said Type (B) radicals ranges from about 40 to 95, the mole percent of said Type (C) radicals ranges from 0 to about 50, the mole percent of said Type (A) radicals ranges from about 5 to 50.

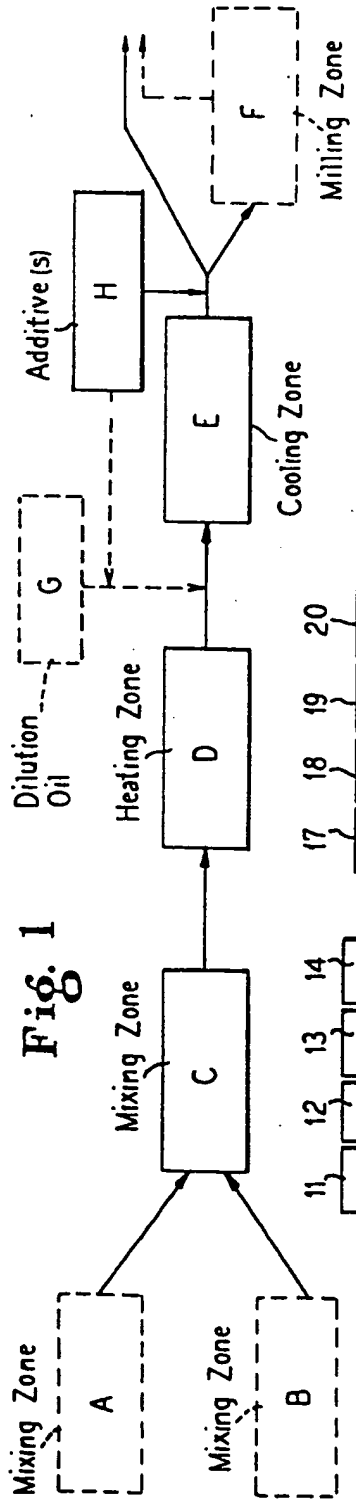


Fig. 1

Fig. 2

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(54) **Grease compositions, oxyaluminum acylate intermediate compositions useful in the preparation thereof, and process for making such.**

(57) Mixed aromatic/aliphatic oxyaluminum acylates are provided which have utility for the manufacture of greases without generating by-product alcohol and without requiring the use of water and wherein the ratio of the number of aromatic radicals to the number of aliphatic radicals ranges from 2:3 to 19:1. Processes are provided for making such compounds including a continuous process. Also premix compositions useful in grease manufacture are provided along with methods for preparing new greases therefrom.

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EUROPEAN SEARCH REPORT

Application number EP 80 10 7252

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Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	
X	DE - B - 1 120 049 (BRITISH PETROLEUM CO. LTD.) * Column 5, line 50 - column 6, line 46; column 1, line 35 - column 3, line 10 *	1-8	C 07 F 5/06 C 10 M 5/12 5/14
X	US - A - 2 599 553 (BRUCE W. HOTTON) * Column 9, line 25 - column 10, line 26; column 5, line 72 - column 6, line 54 *	1-8	
X	US - A - 3 591 505 (ARTHUR T. POSISHUK) * Column 6, line 30 - column 8, line 7; column 4, line 59 - column 5, line 75 *	1-16	TECHNICAL FIELDS SEARCHED (Int. Cl.³) C 07 F 5/00 C 10 M 5/00
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X	US - A - 3 791 972 (WILLIAM A. MYERS) * Column 5, lines 65-75; column 6, lines 4-9; column 4, line 68 - column 5, line 14 *	1-8	CATEGORY OF CITED DOCUMENTS X: particularly relevant A: technological background O: non-written disclosure P: intermediate document T: theory or principle underlying the invention E: conflicting application D: document cited in the application L: citation for other reasons
X	US - A - 3 776 846 (WAYNE W. BAILE) * Column 1, lines 11-27; column 13, lines 6-32 *	1-8	
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X The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
The hague	30-11-1981	SUTER

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EUROPEAN SEARCH REPORT

Application number
EP 80 10 7252
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Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	
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X	GB - A - 825 878 (HARDMAN AND HALDEN LTD.) * Page 7, examples 1,3; page 3, formula 37 * --	1-6	TECHNICAL FIELDS SEARCHED (Int. Cl.³)
D	US - A - 3 345 291 THEODORE H. KAUNDAKIJAN) * Column 4, line 65 - column 6, line 14 * ----	1-6	